Nonlinear Interactions of Dipolar Excitons and Polaritons in MoS_2 Bilayers

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Coherent superpositon of excitons with photons allows for the formation of half-light half-matter bosonic quasiparticles called polaritons. Nonlinear interactions of excitons strongly coupled to light are key for accessing quantum many-body phenomena in polariton systems [1]. Two-dimensional semiconductors provide an attractive platform for the strong light-matter coupling owing to the many controllable excitonic degrees of freedom [2]. Resonant charge tunnelling between two adjacent TMD layer leads to exciton hybridization, which recently emerged as a novel platform for unexplored nonlinear optical phenomena [3]. Here, we employ hybridized inter layer excitons (hIX) in bilayer MoS_2 to realize highly nonlinear excitonic and polaritonic effects. Such interlayer states in MoS_2 possess an out of-plane electric dipole as well as a large oscillator strength, unique among semiconducting atomic homo- and hetero-bilayers [4]. This allows us to realise dipolar polaritons by embedding MoS_2 bilayers in microcavities. We find that under resonant excitation, both hIX and dipolaritons exhibit 10 times stronger nonlinearity compared with excitons and polaritons in monolayer MoS_2. This is further enhanced by a factor of 10 when hIX and intralayer excitons, sharing the common valence band, are excited simultaneously. This provides access to an unprecedented nonlinear regime which we describe theoretically by introducing concepts of *inter-exciton phase space filling* and *hole crowding*. The presented insight in many-body interactions provides new tools for accessing sought-after few-polariton quantum correlations.

References

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Figures

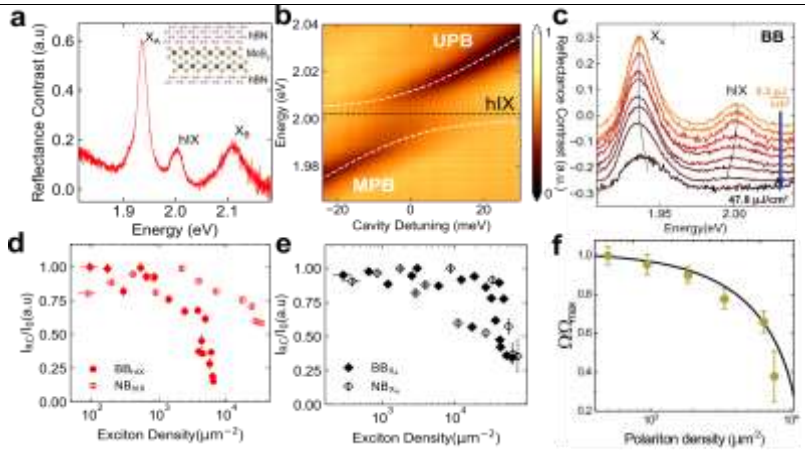

Figure 1: (a)RC spectrum of BL MoS_2 (4K)(b) Strong coupling of hIX-polaritons(c)Fast bleaching of hIX under broadband(BB) excitation, with increasing fluence. This is significantly different under Narrow Band NB excitation(d), in contrast to X_A which shows similar behaviour under the two regimes(e).(f)Large non-linearity of hIX-polaritons shown in the collapse of Rabi splitting, Ω.